

Review ESSD-2023-401, Global N₂O budget

Remarkable compilation of many N-relevant processes. Hard to review, particularly because they have many experts in their author list. Definite merit for ESSD, but needs key improvements.

Basic approach: For important N-molecule, one needs to pay attention to a wide variety of sources, sinks, and biochemical reactions. This paper represents best efforts. They itemize important sources, sinks, and reactions. They draw conclusions by source and by regions. They list abundant uncertainties but rarely in numerical terms. Instead, authors focus on geographic patterns/biases of these uncertainties. They recommend that uncertainties get resolved (“reconciled”) in future observations and from future models.

For examples: Line 1173: “large uncertainty in the estimates of agricultural N₂O emissions”; Line 1209: “large discrepancy in natural soil emissions among NMIP2 models exists”; Line 1391: “variability of these emissions remains uncertain” (referring to emissions from continental shelf regions); etc.

They miss (fail to address) test-able hypothesis: that uncertainties remain so large that conclusions prove speculative (at best). Reader finds zero discussion of cumulative numeric uncertainty. In very practical terms, this reader suggests that uncertainties mapped geographically in Figure 21 overwhelm any regional signals authors might hope to identify in Figure 13.

In GCB, reader immediately (abstract) learns that those authors present all data with consistent specified uncertainty of ± 1 sigma. In global methane budget, reader confronts ranges (min/max) but those authors write (explicitly, in second paragraph of Introduction): uncertainties in regional emissions may reach 40%–60% (of global mean). For N₂O, where obs come directly from same sources as for CH₄, where microbial processes intervene in both sources and sinks, and where models have similar (plus, additional?) weaknesses as for other chemical or transport models, one expects similar quantitative uncertainty info? Should readers assume that we know N₂O concentrations and fluxes with better-than-CH₄ uncertainty, or with worse? Reader here gets no hints. This particular reader assumes ‘worse’! Not because authors have done a poor job (instead, they seem to have done a very good job) but because N₂O represents a more difficult, complicated reactive molecule than e.g. CO₂ or CH₄. If the target remains more elusive, authors must demonstrate and exercise BETTER methods to identify and quantify. Disparaging statement above (e.g. that uncertainties remain so large so as to preclude conclusions on specific sources or assigned to specific regions) represents a clear refutable hypothesis. Unfortunately, authors never pose nor address such a hypothesis.

Validation remains conceptually and quantitatively difficult for these global budgets. Other ESSD products showed good success by using ‘leave-one-out’ techniques. Because these authors report multiple atmospheric data sources, multiple model outcomes, etc., they might choose to pursue similar leave-one-out strategies? Or, and this would prove relevant to uncertainty issues, authors feel reluctant to identify one obs data set or one model as ‘reference’?

I identify many small but necessary changes below. Until, however, readers gain a complete quantitative discussion on uncertainty, I doubt that suggestions that follow pertain. Manuscript needs serious overhaul; small fixes unimportant on that scale.

Line 84: “accumulating in the atmosphere since the pre-industrial period”. No evidence presented here. Most data in this paper start in 1980. No citation for statement, in next sentence, that N₂O concentrations “from 270 parts per billion (ppb) in 1750”. How do we know

pre-1980 N₂O concentrations? In other publications, GCP considers 1750 as start of industrial period? 'Pre-industrial' seems unsupportable and too vague. I know what authors intend here but many ESSD readers will not?

Line 92: if fluxes increased by 40% over 1980 to 2020 but concentrations increased only 25% (from 1750 to 2022, line 85) then something must also have changed in sinks? E.g. Fig 1 shows at least three sinks (downward arrows, including one massive downward arrow); atmospheric concentrations must represent some balance of these processes? Not clear here, nor elsewhere in this manuscript. Authors job to compile and present best data (good on them) but also to explain basic balance / imbalance of global N₂O budget? This reader might understand subtle differences here but many readers will not? Somewhere, in abstract or exec summary, readers need to find concise summary?

Lines 107 to 116: Good summary here! Compares recent work (1980 to 2020) to ice core records. No mention of 'industrial' or 'pre-industrial'. Revise abstract in light of what you have here? Also no mention of stratospheric processes (O₃ impact) or sinks but these processes emerge later? Or, somewhere, a sentence that this budget focuses (necessarily) primarily on tropospheric processes?

Line 110: "It" You mean 'these'? Or, 'these concentrations'? Eliminate this sentence, because you do not need two successive mentions of ice core records?

Here, you use and cite units in fluxes of ppb per year. In abstract and next (emissions) paragraph you instead use Tg N per year. Settle on most useful or most appropriate set of units? Or, include a table that helps readers quickly convert?

Line 195: "most emitted"? Most often emitted? Most emitted by net (atmospheric) concentration or by flux? Most reactive? Need slight clarification here.

Line 198: "mole fractions have increased by more than 25% since the pre-industrial era, from 270 parts per billion (ppb) in 1750 to 336 ppb in 2022" But, figures here only show data since 1980. If increase since pre-industrial values is true (as I accept), reader needs a citation or source for such data?

Line 200: detailed and well-referenced sentence starting 'The 20th century rate' renders the previous sentence moot; reader does not need to see both. ESSD/Copernicus impose some punctuation standard for '20th century'?

Line 204: "growth rate of atmospheric N₂O, the mean annual growth" need changed punctuation here, e.g. growth rate of atmospheric N₂O: the mean annual growth.

Line 217: "Reducing N₂O emissions is a required net-zero greenhouse gas (GHG) emissions and the recovery of stratospheric ozone" Something missing here? Required to meet GHG targets, and to allow (or foster) recovery?

Line 218: Complete "N₂O mitigation measures"? I think Pier's paper pointed out that remaining CO₂ targets/budgets for 2C disappeared into the 'noise' of other GHG mitigation efforts (e.g. for N₂O) but did not address N₂O mitigation impacts directly?

Line 221: "Implementing N₂O mitigation" you already said this in previous sentence. This sentence seems redundant?

Line 223: Nitrification and denitrification might both impact N₂O production but, with one a source and one a sink, they can't both "contribute". Awkward phasing for most readers.

Line 225 and following: good list but punctuation should change to semi-colon between each of 21 factors? Proof readers will know.

Line 225 and following: check Fig 3 to ensure tight correlation with list here, by exact terms and directions of arrows? I think I counted 21 fluxes in Fig 3 but with uncertainty about whether to count bidirectional arrows as one or two terms?

Line 262: But, no red arrow (indirect anthropogenic impact) from coast oceans box of Fig 3?

Line 266: Good list but, strictly speaking, these should not fit in the category of terrestrial natural ecosystems?

Line 284: “multiple BU (BU) and TD (TD) methods” something missing or awkward here?

Line 302: “all possible” but readers just learned that you had to ignore termite sources for lack of data? Perhaps all ‘plausible’. Or, all ‘quantifiable’? Change wording to reflect availability of reliable data? Lists and categories that follow seem reasonable and well-documented. Later (Line 1443) authors devote an entire paragraph to “missing fluxes”. “All possible” remains confusing and/or inappropriate.

Line 316: very important if slightly confusing paragraph. Put this in a table, instead? Ala Table 1 in GCB? Fluxes, change rates of fluxes, atmospheric concentrations: too much for reader to remember without a reference table?

Line 341: ‘are’ rather than “is”?

Line 364: to “to develop” and “quantified” in same sentence? Need some attention to tense here?

Please ensure to define bottom-up (BU) and top-down (TD) once and only once, then attend to all subsequent uses of abbreviations to ensure coherence. E.g. to this reader, text in legend to Figure 4 (line 379) seems confusing?

Here, confusion threatens overall merit of this work. By this point reader has confronted 21 types of N₂O fluxes, recasting of those types into six broader categories, definition of units (fluxes, change rates of fluxes, concentrations), parsing across geographical regions (including to a few specific countries), and - finally (!?) elucidation by 31 (more if one counts same model run at two different spatial resolutions) inventories and global, regional or process models. Huge effort by authors to compile all this! Please keep readers well-informed and cognizant of which source (or sink) estimates apply to which categories. The category ‘Shelf’ for example, which authors intend as one depth-limited region of perimeter oceans, remains confusing as used. Authors will know best what they need to report and how but, unfortunately, present parsing and arrangement implies perfunctory approach while authors prefer to project entire effort as careful and complex. Some better way to convey complexity and uncertainty? Not clear for me. I plead for better overall arrangement or at least an ongoing outline to help readers? Table 1 represents a comprehensive list, without reliability designation? Figure 4 complicates when it might clarify?

Line 415: Copernicus publisher adheres to standard mechanism to handle ‘submitted’ references. Not this one, unfortunately; authors and editors need to correct.

Line 417: Which “observation-based analysis”?

Line 423: percent of what?

Line 424: “Shelf processes” in Table 1 actually represent “continental shelves” as specified here?

Line 434: If authors already listed all pertinent (hi-res) ocean biogeochemistry models in Table 1, does reader need a second list here? Again, this reader wishes for some clarification: unable to assign priority to any one model, authors have chosen to use them all, to use a mean, to use a median? How do uncertainties from individual models penetrate into overall global estimates?

Line 455: First reports on deriving uncertainty info from source materials? Reader of ESSD needs more of the same?

Line 465: More useful to list FAO general reports first, then to deal with FAOSTAT specifics on fire types after? E.g. helpful to readers to change order of last and next-to-last sentences?

Line 470 and following: In this section authors need to include continental shelves in a more-general ‘coastal’ category? Needs explicit mention/explanation?

Line 481: not clear where “56%” comes from?

Line 484: “low” or “act as a sink”. Should reader assume you included these sources/sinks or ignored them as insignificant?

Line 486: “SH1-SH7” and line 487 “SH1-SH8” From supplement reader learns that SH1 etc. represent set-up parameters for models involved in NMIP2 but readers need that information sooner, e.g. here?

Line 491: “book-keeping” approach may account for deforestation/reforestation but you lost readers on broader issue of overall issue of land-cover changes on indirect (perturbation?) emissions?

Line 495 and following: Not clear what authors concluded here: should they use older estimate for NH₃ oxidation and lightning production or do they ignore these processes as “small” and inconsistent or unquantifiable?

Line 515: ppb to Tg conversion factor buried here, should appear more prominently in a ‘units’ table?

Line 517 and following: Did authors use $\pm 1.4\%$ uncertainty or IPCC AR5 uncertainty? One applies to concentrations and other to concentration changes? Not clear to this reader.

Line 524: another example of a process (tropospheric loss) too small to appear in overall N₂O budget?

Line 533: ‘is’ rather than “as”?

Line 548: additional uncertainties introduced by these interpolation or re-gridding steps?

Line 553: Back at line 319, we read “Unless specified, uncertainties are reported in brackets as minimum and maximum values of all estimates”. Data presented here (e.g. 315.8 (“315.5-316.2) ppb in 2000 to 335.9 (335.6-336.1) ppb in 2022”) follow this convention? Values in parentheses represent min and max? But, each source (e.g. NOAA, CSIRO) will have gone to lot of effort to

identify uncertainties of their N₂O measurements, reported not as \pm min/max. Min/max tells readers very little about distributions or uncertainties? Please can authors adopt, and adhere consistently to, better more reliable more informative uncertainties?

Line 556: “was” implies singular but this sentence refers two (plural) years, 2020 and 2021?

Line 556: “30% higher than the average value in the decade of the 2010s” Not sure that readers can confirm this information from Figure 2? Inset of Figure 2, which purports to show annual growth rate, has no uncertainties, no demarcation of decades, nothing to help readers follow (or, dispute) authors’ conclusions. Help, please.

Line 563, Figure 5: No uncertainties in obs nor in model outcomes?

Line 571: “with large uncertainties (Figure 6).” What uncertainties? \pm min/max as above? Something different here? No information! Nothing in panels or figure legend?

Line 579, Figure 6: Except for panel C (Other direct anthropogenic emissions), all uncertainties (if shown by color ranges) appear to increase, 1980 to 2020? Not a positive report? Should readers assume that capability to construct N₂O budget decreases, because uncertainties increase? Not what this reviewer would have expected as measurements and models all improve? If end of 2020 total of 6.7, with range of 3.3 (minimum or ?) to 10.9 (maximum or ?), how can reader trust anything that follows about sectors or regions? If authors expect readers to accept time-dependent changes in N₂O sources or sinks, those readers will need to trust authors’ handling of received as well as generated uncertainties. No evidence provided here.

Line 583 and following, sections on agriculture, other direct, etc.: Lots of work here, compiling and reporting data from trusted sources NIMP2, EDGAR, FAOSTAT, etc., but not one mention of uncertainties. Do all these sources produce ‘perfect’ data? I know, and readers will know better, that each source spends a great deal of time and effort to identify and report uncertainties. All of that effort and info lost here? Not one error bar or uncertainty envelop in any of these figures? Authors need to provide readers a basis to trust these conclusions, to respect authors’ good efforts, but nothing presented here provokes nor supports such respect. Fossil fuel N₂O emissions have remained unchanged for four decades? Weakness in reporting? True? How can any reader know? We need to trust authors to provide explanation with documentation and uncertainties! Instead, nothing provided!

Line 633, “both DLEM and book-keeping approach suggested increasing uncertainties in post-deforestation pulse effect”. Really? Increasing uncertainties? Expressed as min/max, 95CI, per cent of total, \pm x sigma, what? If authors want readers to accept this contention about perturbation fluxes, those readers will want to have seen consistent approach to uncertainties up to this point and will need more details here.

Lines 648 and following: Reader would like to accept that natural N₂O fluxes have not changed over four decades but a) this reader doubts that contention and b) authors have given no indication of their skill or knowledge to back up such contentions.

Line 650 and following: do authors now (or, again) present minimum and maximum values? Doubtful, but no information given to help readers decide? Readers would like to accept authors’ contentions on all these natural N₂O fluxes but, without uncertainty information and specifications, how can we?

Line 685, TD estimates: Inversions, as published, include substantial uncertainties. As presented (plotted) here, however, readers get no indication of authors expertise at assimilating

and assessing TD info. No error bars on any plot, can't be true? What valid signals emerge from what noise? No sense of that provided in this section.

Line 778, Figure 13: Total N₂O emissions (panel A), look nothing like panel A of Figure 6 (total anthropogenic). Total here represents Fig 6 plus non-varying natural total of 12 (Line 651)? 7 plus 12 gives 19, perhaps within uncertainty (?) of this Figure? If this reader has made too-simple assumptions or additions, authors have not given sufficient information to prevent my errors? What do uncertainties shown here represent? Cumulative for BU and TD? From where? If we can't understand panel A, why should any reader give any credence to panels b to t? If I calculate instead from numbers given in Table 2 (again, with min and max?), I get 18 BU or 17 TD total N₂O sources, minus stratospheric sink of 13 or total atmospheric sink of 14 to get a residual in atmosphere of 3 to 5? Compared to 6 as provided authors? Sorry, does not compute correctly. In notes to Table 2 one reads that uncertainties follow AR5 with detailed notes provided in Supplement. Sorry again, this reader will not comb through supplement to elucidate uncertainty info. Many readers will simply give up at this point.

Lines 790 to 1084, regional extrapolations. More hard work by authors but basically bogus without uncertainties or some mechanism to validate?